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# DESIGN IMPROVEMENTS FOR CARBON-14 CRYOGENIC DISTILLATION SYSTEM

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Cryogenic distillation of carbon monoxide has been shown to be an effective way to concentrate carbon-14 wastes stripped from ion exchange resins used in CANDU reactors. This report describes design improvements to the demonstration column that were implemented during column testing. The newly designed cryogenic distillation system will be a key component of the $^{14}C$ pilot plant to be constructed at Research Division.

## 1. INTRODUCTION

A proposal has been made to New Business Ventures Division to build a pilot plant at Research Division to demonstrate the recovery of carbon-14 from nuclear power plant in exchange resins and to also demonstrate the enrichment of this carbon-14 to 99%. At this concentration level, carbon-14 is a valuable commercial product worth millions of dollars per year. The New Business Ventures Management Board has provisionally accepted this proposal. Construction of the pilot plant will go ahead when the new Research Division Radiation Laboratory is complete and when minority funding of this project by potential carbon-14 customers is in place.

The enrichment of carbon-14 is done by cryogenic distillation of carbon-monoxide. For the past several years, Research Division has studied carbon-14 enrichment using carbon-13 distillation. Prior to the operation of this distillation equipment, a report describing its operation was issued/1/. This report updates the previous report giving the design improvements that were implemented after the start-up of experiments. A description of these changes will assist the pilot plant designers.

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## 2. DESIGN CHANGES

A number of major changes have been made to the carbon-14 cryogenic distillation system since the August 15, 1985 report /1/. That report described the design and construction of this system prior to any experimental testing. During testing several changes and improvements were made. The changes are reported here with the same format as reference 1 so that the two can be easily compared.

The Design, Construction and Assembly section (Appendix 1) describes details of the new column and packing.

The Carbon Monoxide Handling System (Appendix 2) shows changes made which allow both total reflux operation, as previously used, as well as the capability of the automatic control of material streams to and from the distillation unit/2/. This unit permits continual (pulsed) feed of carbon monoxide to the still and continual (pulsed) withdrawal of an enriched $^{13}C$ or $^{14}C$ and a $^{13}C$ or $^{14}C$ depleted stream of CO.

Changes to the Liquid Nitrogen System (Appendix 3) were necessary because of the addition of a new 6000 litre liquid nitrogen supply tank which operates at almost 4 times the pressure of our previous storage tank/3,4/.

Changes to the high vacuum system listed under the Insulation section (Appendix 4) have resulted in lower operating pressures ($2x10^{-9}$torr) in the insulating high vacuum which surrounds the distillation column.

The Monitor and Control Systems (Appendix 5) were changed so that we could more closely monitor pressures in the distillation unit.

The section on Safety Systems (Appendix 6) describes the addition of a new oxygen monitor as well as a new carbon monoxide monitor which are both situated in the lab. Additional carbon monoxide over pressure instrumentation has also been added to prevent escape of any CO outside of the laboratory in the event of loss of liquid nitrogen cooling. Reboiler power control has been added which is activated when a low liquid nitrogen level is reached. This conserves the amount of nitrogen remaining in the lower liquid nitrogen reservoir and increases the length of time following loss of coolant supply, before CO has to be vented to the CO storage tank.

Appendix 7 lists some of the cryostill's typical operating parameters.

## 3. SUMMARY

Cryogenic distillation of carbon monoxide is an effective way to enrich carbon-14. Several design improvements have been made to the enrichment hardware since the initial report on carbon-14 enrichment was issued/1/. This report summarizes the changes that were made during the testing of our cryogenic distillation system. This information is required for the detailed design of a carbon-14 pilot plant.

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2. F.E. Bartoszek, "Automatic Control of Material Streams in the Research Division Carbon-14 Cryogenic Still". Research Report No 86-219-H.

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4. R. Scheer, Research Division Report, in preparation.

## APPENDIX

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## APPENDIX 1

## DESIGN CONSTRUCTION AND ASSEMBLY

The test apparatus described in reference one can be fitted with an entirely new column in about 3 weeks. The column that has performed best for CO distillation was first installed in March 1986, and is currently being used for testing the automated carbon monoxide feed system, and product and waste withdrawal systems. This column contains 395.04 grams of Helipak "B", #3013, which is a stainless steel packing measuring 0.050"x0.100"x0.100". The new packed column length is 148.4 cm.

## APPENDIX 2

## CARBON MONOXIDE HANDLING SYSTEM

The 1/16" O.D.(0.010" I.D.) stainless steel connecting lines leading to the reboiler (2 lines) and to the column (feed line for the automated material stream) have been replaced with 1/16" O.D. (0.030" I.D.) stainless steel tubing. The larger internal diameter tubing was used to reduce the pressure drop in the connecting tubing allowing the pressure gauges to more quickly and accurately reflect actual conditions in the distillation unit. The larger lines also help to increase the rate at which feed and withdrawal can take place.

The 1/16" O.D. tubing which joins valve 7 (Figure 1) to the bottom of the reboiler remains at a length of 274 cm.

### 1.0 CO FEED

The addition of the automated material stream controller reported in /2/ required the replacement of the single-stage regulator with a two-stage vacuum regulator model # 3102-350. This permits feed of CO to the still at delivery pressures below 1000 torr. Initial charging of the distillation unit with CO is the same as reported in /1/.

The 1/16" stainless steel tubing (0.030" I.D.) connected between valve 9 and to a point 28.1 cm below the top of the packing remains at a length of 129 cm (Figure 1). The middle sample port, connected between valves 5 and 9, was previously used for sampling during total reflux operations. A length of 1/8" O.D. copper tubing now connects to solenoid SM1 (Figure 2) and is now used for the CO feed stream.

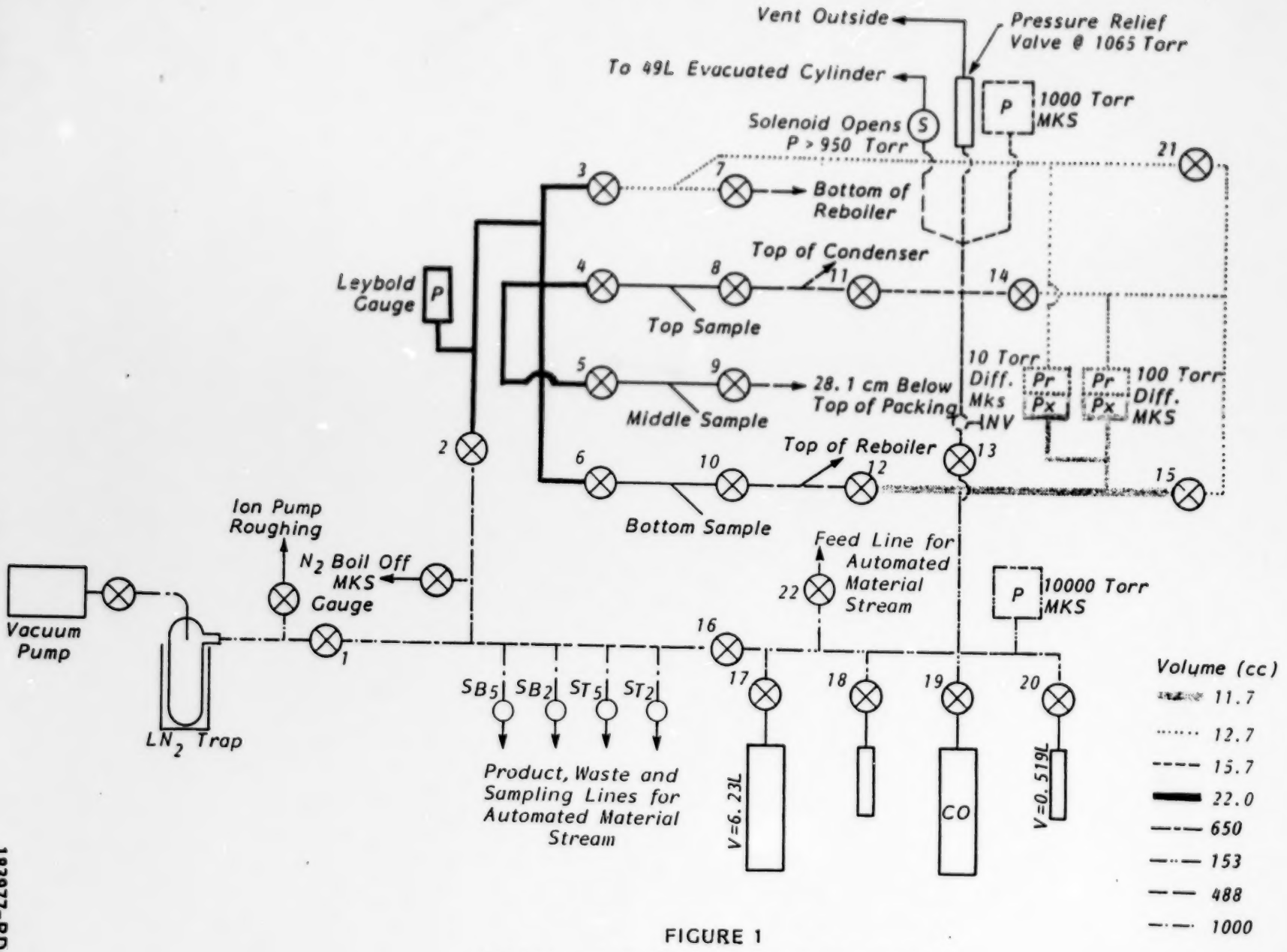



FIGURE 1

CO FEED PLUS GAS SAMPLING SCHEMATIC
(Revision of Figure 5 in Reference 1)

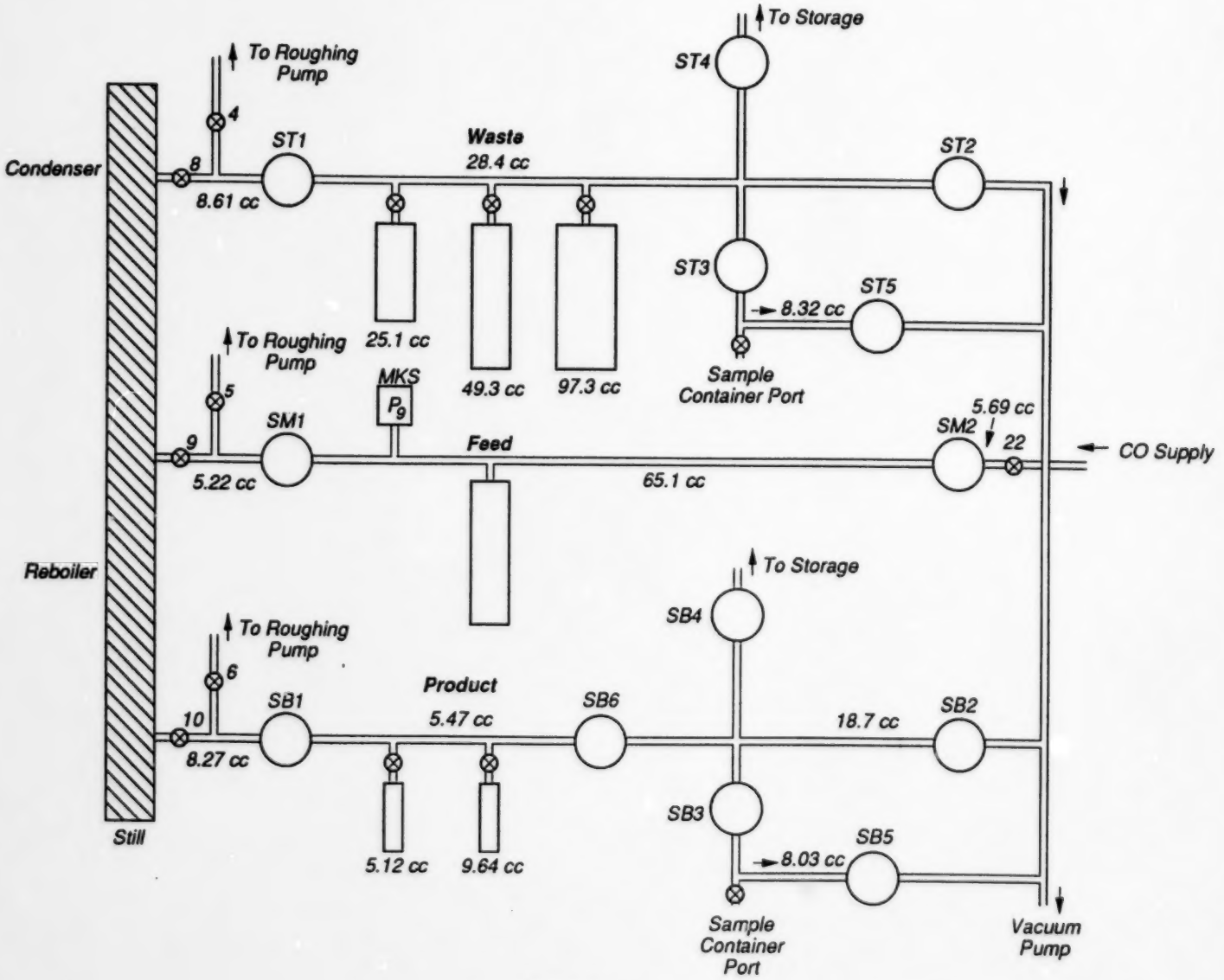



**FIGURE 2**

**SCHEMATIC DIAGRAM OF THE LAYOUT OF THE SOLENOID VALVES TO THE CRYOSTILL**
**(Revision of Figure 1 in Reference 2)**

## 2.0 PRODUCT AND WASTE REMOVAL

The location of the sampling manifold was changed to accommodate the addition of the automated material stream controller. The top sample port of Figure 1 is connected to the valve ST1 in Figure 2 by a 1/8" O.D. copper tube. This is the location of the waste removal from the distillation unit. Gas sampling for isotopic analysis purposes can also be taken at this location.

The 1/16" tubing connected between valves 10 and 12 and the top of the reboiler remains at 250 cm long. The bottom sample port connected between valves 6 and 10 (Figure 1) is now connected to solenoid SB1 (Figure 2) via a 1/8" O.D. copper tube. This is where the product withdrawal stream exits the distillation unit. This is also the location for product sampling for isotopic purposes.

During total reflux operation of the cryostill, sampling involves manual switching of various solenoids. Listed below is the procedure for taking top and bottom samples. Refer to Figures 1 and 2.

Top sampling procedure (volume of gas removed = 45.62 cc)

1. Connect sample tube then open stopcock and sample tube manual valve.
2. Plug in shorting plugs.
3. Open ST1, ST2, ST3, ST5 and evacuate until the pressure is $10^{-3}$ torr then close ST1.
4. Close valve 11.
5. Open valve 8 for 30 seconds then close it.
6. Close ST2, ST3, ST5.
7. Open valve 4 until the pressure is $10^{-3}$ torr then close it.
8. Open ST1.
9. Open valve 8 for 30 seconds then close it.
10. Open valve 11 and ST3.
11. Close sample tube stopcock.
12. Open ST2 and ST5 and evacuate until the pressure is $10^{-3}$ torr.
13. Close ST1, ST2, ST3, ST5, and sample tube manual valve.
14. Unplug shorting plug.
15. Remove the sample tube.

Bottom sampling procedure (volume of gas removed = 40.71 cc)

1. Connect sample tube then open stopcock and sample tube manual valve.
2. Plug in shorting plugs.
3. Open SB1, SB2, SB3, SB5, SB6 and evacuate until the pressure is $10^{-3}$ torr then close SB1.
4. Close valve 12.
5. Open valve 10 for 30 seconds then close it.

6. Close SB2, SB3 and SB5.
7. Open valve 6 until the pressure is $10^{-3}$ torr then close it.
8. Open SB1.
9. Open valve 10 for 30 seconds then close it.
10. Open valve 12 and SB3.
11. Close sample tube stopcock.
12. Open SB2 and SB5 and evacuate until the pressure is $10^{-3}$ torr.
13. Close SB1, SB2, SB3, SB5, SB6 and sample tube manual valve.
14. Unplug shorting plug.
15. Remove sample tube.

During operation of the feed system and the product and waste withdrawal systems sampling is a semi-automatic procedure. Listed below is the procedure for taking top and bottom samples simultaneously.

1. Connect sample tubes and open sample tube manual valves.
2. Plug in shorting plugs.
3. Turn on sample container pumpout alarm and open ST5 and SB5 during pump/fill cycle.
4. Open sample tube stopcocks and evacuate until the pressure is $10^{-3}$ torr.
5. Close ST5 and SB5.
   Note: For no product withdrawal operation - Open valve 10, let controller operate for 2 feed/withdrawal cycles.
6. Change SB6 from switchable to always on position and push sample select button during pump/fill cycle.
7. During the next feed/withdrawal cycle close sample tube stopcocks.
   Note: This has to be done before the next pump/fill cycle so that none of the sample is pumped away.
8. Wait for the next feed/withdrawal cycle then close SB6.
   Note: For no product withdrawal operation - close valve 10.
9. Open ST5 and SB5.
10. Close ST5 and SB5 when the pressure is $10^{-3}$ torr and turn off sample container pumpout alarm.
11. Close sample tube manual valves.
12. Unplug shorting plugs.

## APPENDIX 3

## LIQUID NITROGEN SYSTEM

### 1.0 PRESSURE CONTROL

The cartesian manostat which controlled the pressure above the liquid nitrogen was replaced by an MKS 300 position butterfly valve controller model # 253A-20-40-TSP (Figure 3, valve $S_C$). The addition of the controller made the 45.6 $\ell$ ballast volume unnecessary so it was removed.

For experiments with the liquid nitrogen ($LN_2$) at or above atmospheric pressure, the vacuum pumps and oil filter can be bypassed. The exit of valve $S_C$ directly connects to the 3-way solenoid $S_B$ and an additional normally closed ASCO solenoid. This additional solenoid will be opened only during the pump down and dump cycles of the $LN_2$ filling cycle to allow for extra venting of the nitrogen gas.

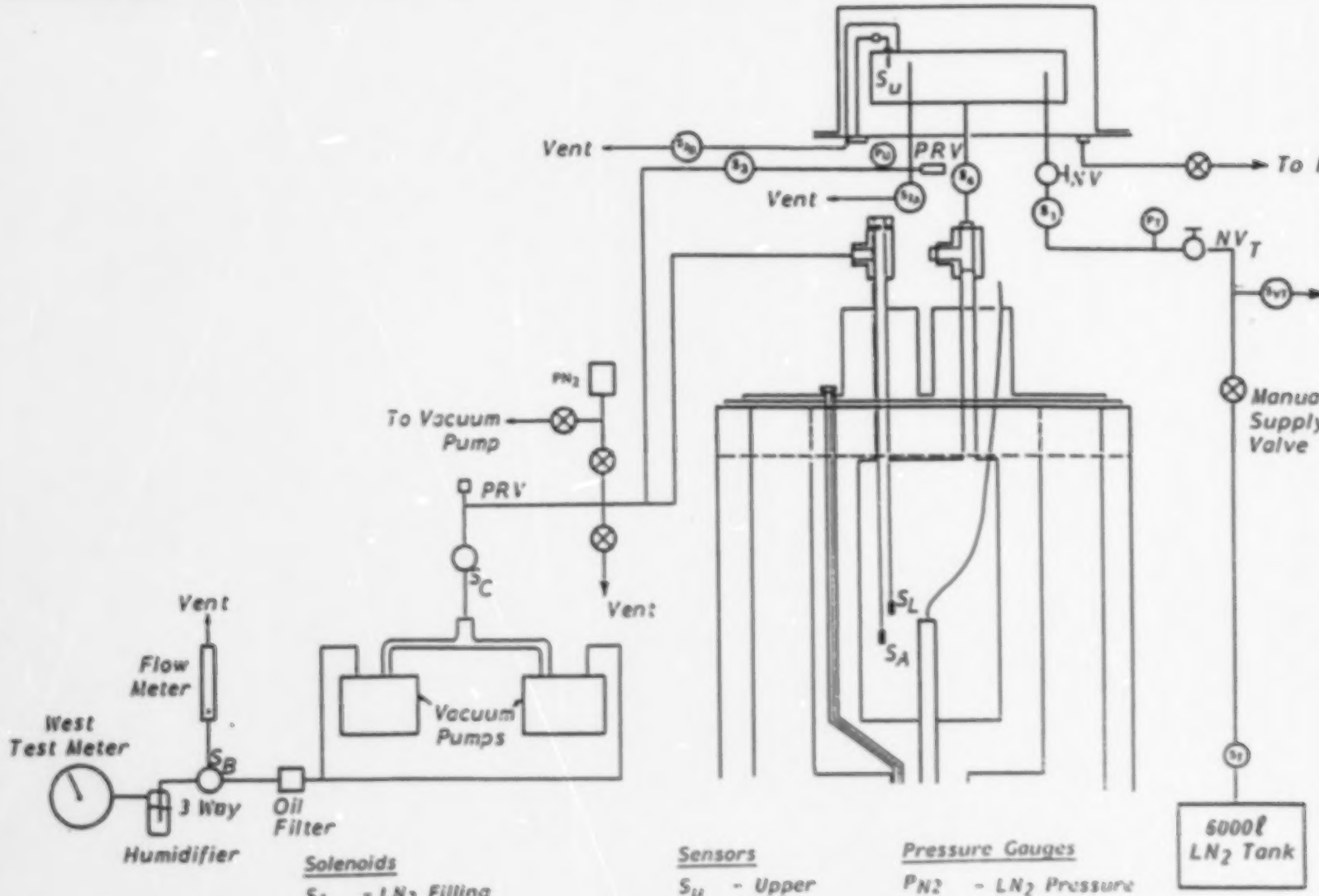
### 2.0 PRESSURE RELIEF

The greatest effort has gone to changing the liquid nitrogen delivery system to accommodate the higher pressure (90 psi) in the long transfer lines that connect the cryostill to the 6000 litre liquid nitrogen storage tank situated outside the building. The changes that had to be made to the valves and lines of the delivery system can be seen in Figure 3. A second vent line, controlled by a 1/2" normally closed ASCO solenoid $S_{2B}$, had to be added because there was insufficient venting to allow quick filling times. The new 1/2" O.D. stainless steel tubing vent line provides enough venting when opened in parallel with the earlier 1/4" line ($S_{2A}$). Another 1/2" feed line vent, controlled by a 1/2" normally opened Magnotrol solenoid ($S_{VT}$), was added immediately after the manual $LN_2$ supply tank valve to cool down the lines that connect the outside tank to the cryostill.

To reduce the pressure of the liquid nitrogen before it enters the upper reservoir a needle valve ($NV_T$) was added before solenoid $S_1$. At present NV is set at 1.7 and $NV_T$ is at 1.5. Gauges were added, ($P_T$,$P_U$) to monitor pressures in important locations and can also be used for trouble-shooting.

### 3.0 AUTOMATIC CONTROL SYSTEM FOR LIQUID NITROGEN TRANSFER

Pump-down and dump times were changed on the $LN_2$ fill-timer box reported in /3/ to 45 seconds and 23 minutes respectively. A timer was added to the Torr-Vac autofiller (setting=0.40) to eliminate the effect of liquid nitrogen splashing the sensor in the upper reservoir. The splashing would result in inadequate fills because the sensor trips prematurely when it is splashed.




**Solenoids**

| | |
|---|---|
| $S_1$ | - $LN_2$ Filling |
| $S_{2A}$ | - ¼" Vent |
| $S_{2B}$ | - ¼" Vent |
| $S_3$ | - Vacuum Pump |
| $S_4$ | - Still Filling |
| $S_C$ | - Pressure Control |
| $S_B$ | - Wet Test Meter Bypass |
| $S_{VT}$ | - $LN_2$ Feed Line Vent |
| $S_T$ | - $LN_2$ Main Supply Tank |

**Sensors**

| | |
|---|---|
| $S_u$ | - Upper |
| $S_L$ | - Lower |
| $S_A$ | - Low Alarm |

**Pressure Gauges**

| | |
|---|---|
| $P_{N2}$ | - $LN_2$ Pressure Control |
| $P_T$ | - Tank Pressure |
| $P_U$ | - Upper Reservoir Pressure |

FIGURE 3

$LN_2$ SYSTEM

(Revision of Figure 6 in Reference 2)

## 4.0 LIQUID NITROGEN SUPPLY TANK CONTROLLER BOX

Since the cryostill is not the only user of the outside tank, a Liquid Nitrogen Controller box was designed and built/4/. This control box oversees the opening and closing of the main solenoid $S_T$ which is located immediately after the outside tank (Figure 3). This solenoid remains open for a maximum preset time between 1 and 60 minutes. This controller also controls the length of time that the vent solenoid ($S_{VT}$) is open. This time is adjustable from 0 to 6 minutes with the current setting set at 2 minutes and 51 seconds.

## 5.0 UPPER $LN_2$ RESERVOIR

Two 1/2" holes (Figure 4, 5) were drilled into the top reservoir to increase venting capacity. This was required when we switched from a 28 psi $LN_2$ supply inside the lab to a 90 psi $LN_2$ supply outside the building. One hole was drilled through the top wall of the inner chamber and the other was drilled through the bottom flange. A 1/2" O.D., 0.049" wall stainless steel tube connects to the top of the inner chamber, passes through the bottom flange and extends 1" past the flange. Both welds were made on the high vacuum side.

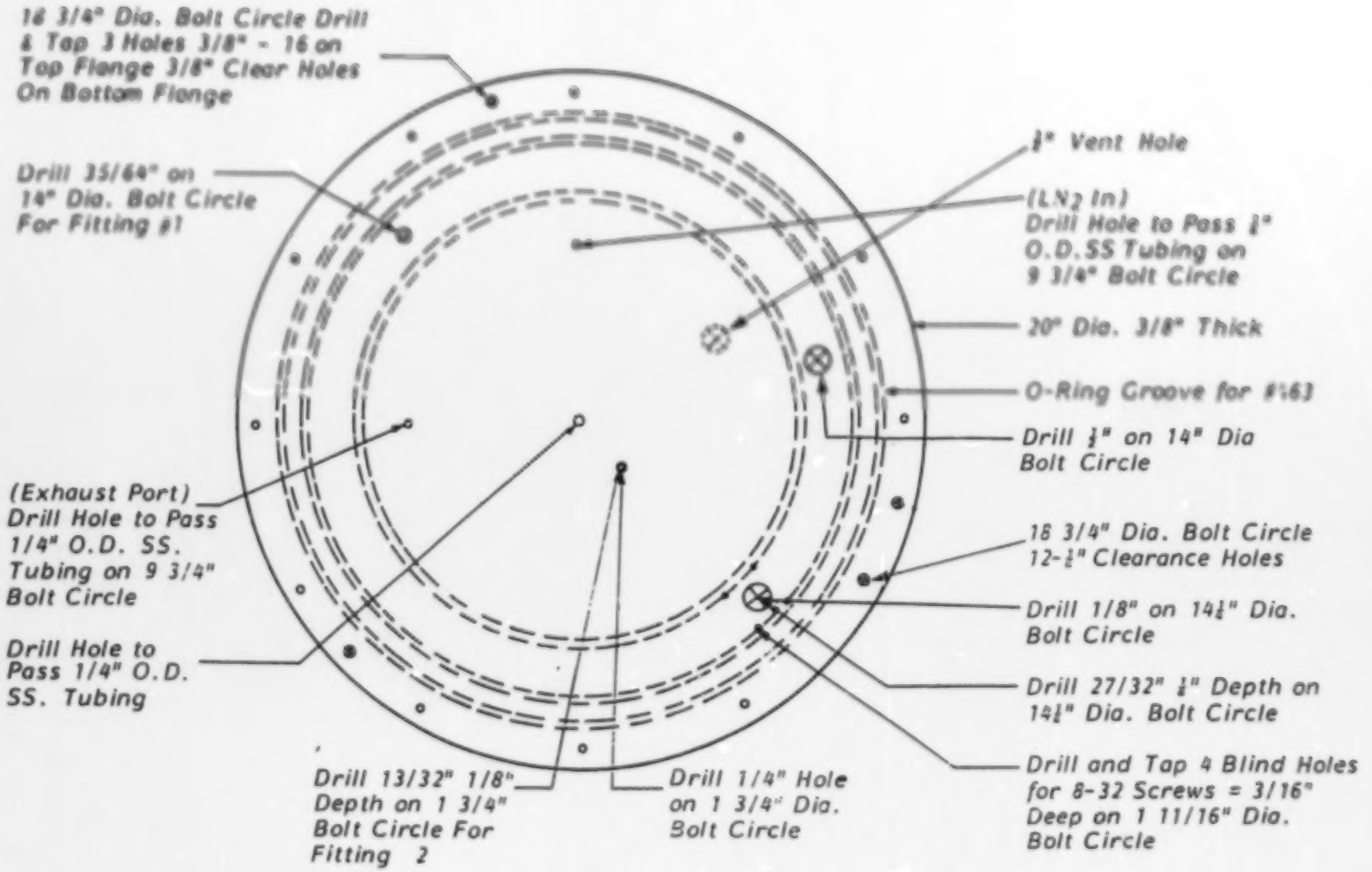
The high vacuum chamber, surrounding the upper $LN_2$ reservoir is now directly connected to the ion pump via a 3/8" O.D. copper tube.

# APPENDIX 4

# INSULATION

All buna-N o-rings in the high vacuum section were replaced with viton o-rings. This helped reduce the operating pressure in the high vacuum to $3 \times 10^{-9}$ torr.

The 220 l/s VacIon Pump required routine refurbishing after 3 years use. It was replaced with a 110 l/s pump of the same make. This size pump has proved to be entirely adequate for our high vacuum requirements.


18 3/4" Dia. Bolt Circle Drill & Tap 3 Holes 3/8" - 16 on Top Flange 3/8" Clear Holes On Bottom Flange
Drill 35/64" on 14" Dia. Bolt Circle For Fitting #1
(Exhaust Port) Drill Hole to Pass 1/4" O.D. SS. Tubing on 9 3/4" Bolt Circle
Drill Hole to Pass 1/4" O.D. SS. Tubing
Drill 13/32" 1/8" Depth on 1 3/4" Bolt Circle For Fitting 2
Drill 1/4" Hole on 1 3/4" Dia. Bolt Circle
Drill and Tap 4 Blind Holes for 8-32 Screws = 3/16" Deep on 1 11/16" Dia. Bolt Circle
Drill 27/32" 1/4" Depth on 14 1/4" Dia. Bolt Circle
Drill 1/8" on 14 1/4" Dia. Bolt Circle
18 3/4" Dia. Bolt Circle 12-1/4" Clearance Holes
Drill 1/4" on 14" Dia Bolt Circle
O-Ring Groove for #163
20" Dia. 3/8" Thick
(LN2 In) Drill Hole to Pass 1/4" O.D. SS Tubing on 9 3/4" Bolt Circle
1/4" Vent Hole

Scale 1/4" = 1"

FIGURE 4

UPPER LIQUID NITROGEN RESERVOIR FOR
CRYOGENIC DISTILLATION APPARATUS (TOP VIEW)
(Revision of Figure 15 in Reference 1)

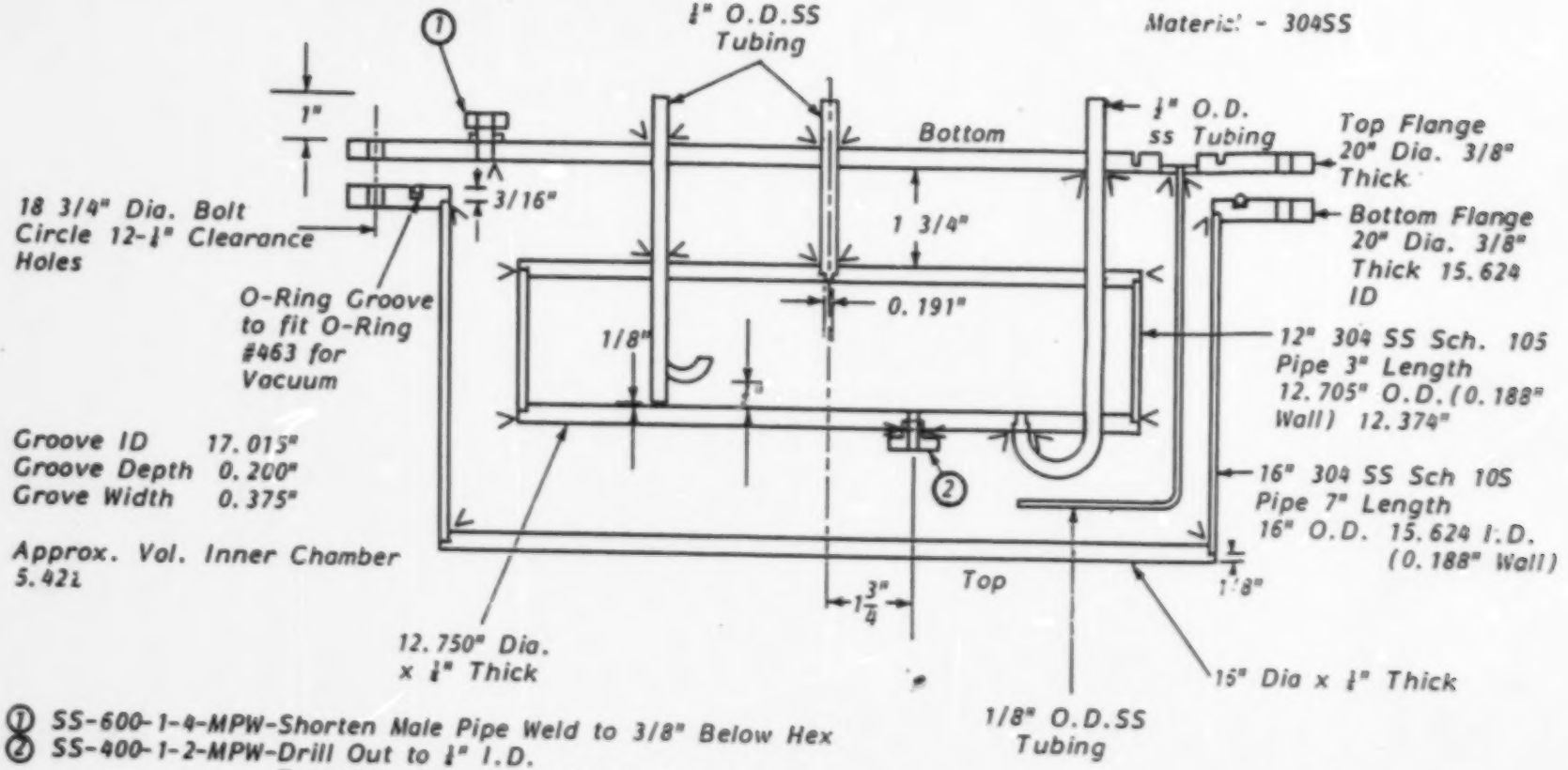

1
1" O.D.SS Tubing
Material - 304SS
1"
3/16"
18 3/4" Dia. Bolt Circle 12-1/4" Clearance Holes
O-Ring Groove to fit O-Ring #463 for Vacuum
Groove ID 17.015"
Groove Depth 0.200"
Grove Width 0.375"
Approx. Vol. Inner Chamber 5.42ℓ
12.750" Dia. x 1/4" Thick
Bottom
1/4" O.D. ss Tubing
Top Flange 20" Dia. 3/8" Thick
1 3/4"
0.191"
1/8"
Bottom Flange 20" Dia. 3/8" Thick 15.624 ID
12" 304 SS Sch. 10S Pipe 3" Length 12.705" O.D. (0.188" Wall) 12.374"
16" 304 SS Sch 10S Pipe 7" Length 16" O.D. 15.624 I.D. (0.188" Wall)
2
Top
1 3/4
1 1/8"
15" Dia x 1/4" Thick
1/8" O.D.SS Tubing
1 SS-600-1-4-MPW-Shorten Male Pipe Weld to 3/8" Below Hex
2 SS-400-1-2-MPW-Drill Out to 1/4" I.D.
-Turn Hex Down to 0.403" O.D. x 0.25" Long
Scale 1/2" = 1"

FIGURE 5

UPPER LIQUID NITROGEN RESERVOIR FOR CRYOGENIC DISTILLATION APPARATUS (SIDE VIEW)

(Revision of Figure 16 in Reference 1)

## APPENDIX 5

## MONITOR AND CONTROL SYSTEMS

To calculate how much feed is added to the column during feed and product and waste withdrawal, a 1000 torr MKS Baratron pressure transducer (Pg) was installed on the feed line (Figure 2) . The height of liquid CO in the reboiler is now monitored by a 10 torr differential pressure gauge (Figure 1). Previously, the 100 torr differential gauge monitored this pressure and the pressure drop across the column. However, this setup required manual switching of a valve to switch between monitoring the reboiler and column pressures.

## APPENDIX 6

## SAFETY SYSTEMS

### 1.0 $LN_2$ SYSTEM

A new oxygen monitor, model #335 from Teledyne Analytical Instruments measures the percent oxygen in the room. Caution and danger indicators light up at 19% and 17.5% oxygen respectively. The monitor also triggers an audible and visual alarm at these oxygen levels. This monitor warns lab workers when nitrogen venting is starting to displace the oxygen in the lab.

### 2.0 CO SYSTEM

After two failures of the Ecolyser CO monitor's sensor, it was no longer used. The lab CO level is now monitored by a Gas Master model # GSM4 with a range of 0 to 999 ppm CO. At a level of 35 ppm CO the monitor triggers an audible alarm.

The 1000 torr pressure transducer that monitors the condenser pressure activates a normally closed 1/4" ASCO solenoid valve when the pressure exceeds 961 ± 31 torr (Figure 1). When this pressure is exceeded a light indicates that the ASCO valve has been energized allowing the overflowing gas to vent to a 49 litre evacuated cylinder. If the pressure is relieved to below 930 ± 10 torr, the ASCO valve is closed but the indicator remains lit until it is shut off manually.

### 3.0 HEATER POWER CONTROL

Both reboiler heater elements are connected to a reboiler power relay that will cut power to the reboiler when the $LN_2$ alarm sensor gets warm. This means that the lower liquid nitrogen

reservoir is down to 1/2 full and indicates a problem with the $LN_2$ filling system. Cutting power to the reboiler gives the operator more time to react to the problem with the $LN_2$ filling system.

## APPENDIX 7

## TYPICAL OPERATING CONDITIONS

With a CO inventory of 1.83 moles, $LN_2$ pressure of 760 torr, and reboiler power at 16 watts the following operating conditions have been recorded during total reflux operation.

| PRESSURE GAUGES | | FILL-TIMER BOX | | TEMPERATURE CONTROLLER | | TEMP. CORRECTED USING CALIBRATION |
|---|---|---|---|---|---|---|
| $PLN_2$ | 778 torr | Dump timer | 23 min | Diode 1 | 80.90 K | 80.00 K |
| $\Delta P_{REB}$ | 5.7 torr | Pump-down timer | 45 sec | Diode 2 | 80.70 K | 81.00 K |
| $P_{COND}$ | 615 torr | $LN_2$ supply timer | 75 min | Diode 3 | 82.05 K | 82.15 K |
| $\Delta P_{COL}$ | 13 torr | Manual Override | Off | Diode 4 | 81.40 K | 81.20 K |
| | | | | Diode 5 | 81.35 K | 81.25 K |
| | | | | Diode 6 | 81.45 K | 81.35 K |
| | | | | Set Point | 81.60 K | |